AD-A156 243   OPTICAL PROPERTIES OF HETEROSTRUCTURES AND   1/1
               SUPERLATTICES(U) MASSACHUSETTS INST OF TECH CAMBRIDGE
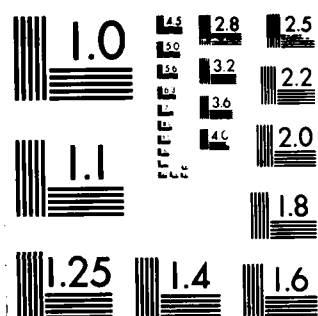   FRANCIS BITTER NATIONAL MAGNET LAB B LAX MAY 85
UNCLASSIFIED   N00014-84-K-0431   F/G 20/12   NL

END
DATE
FILMED
8 -85
DTIC

MICROCOPY RESOLUTION TEST CHART

NATIONAL BUREAU OF STANDARDS 1963 A

OFFICE OF NAVAL RESEARCH

AD-A156 243

ANNUAL REPORT

for

15 JUNE 1984 THROUGH 14 JUNE 1985

for

CONTRACT N00014-84-K-0431

"OPTICAL PROPERTIES OF HETEROSTRUCTURES AND SUPERLATTICES"

DTIC
ELECTE
JUL 8 1985
S   D
          B

Professor Benjamin Lax
Principal Investigator

Massachusetts Institute of Technology
77 Massachusetts Avenue
Cambridge, MA 02139

May, 1985

85 6 3 121

REPORT DOCUMENTATION PAGE	READ INSTRUCTIONS BEFORE COMPLETING FORM

1. REPORT NUMBER	2. GOVT ACCESSION NO. *A156293*	3. RECIPIENT'S CATALOG NUMBER

4. TITLE (and Subtitle)	5. TYPE OF REPORT & PERIOD COVERED
Annual Report	~~Interim~~ 15 June 1984–14 June 1985
	6. PERFORMING ORG. REPORT NUMBER

7. AUTHOR(s)	8. CONTRACT OR GRANT NUMBER(s)
Benjamin Lax	N00014-84-K-0431

9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
Francis Bitter National Magnet Laboratory, MIT Cambridge, MA 02139	

11. CONTROLLING OFFICE NAME AND ADDRESS	12. REPORT DATE
Office of Naval Research Arlington, VA 22219	May 1985
	13. NUMBER OF PAGES 9

14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)	15. SECURITY CLASS. (of this report)
	Unclassified, Unlimited
	15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

DISTRIBUTION STATEMENT A
Approved for public release
Distribution Unlimited

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Keywords

Exciton,   Infrared detector,
Luminescence,   Multiple quantum wells,
Magneto-optics,
Semiconductor lasers, ← end

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

TABLE OF CONTENTS

I. Research Accomplishments

The investigation of the optical properties of hetero-structures and quantum wells in semiconductors has produced a number of results of basic and practical importance. Experimental and theoretical studies of luminescence of a single quantum well under laser excitation established exciton transport as the mechanism for providing the electron-hole pairs which recombine within the well. This contradicts previous models of electron hole transport into quantum wells. The significance to heterojunction and quantum well lasers is obviously relevant.

Another experiment on multiple quantum wells involving selective excitation with tunable pulse dye lasers near the lowest exciton level has led to the discovery of a new lowest excitonic-like state unique to such a two-dimensional (2D) systems. The luminescence characteristic of this lower state has a threshold and persists for a decade of excitation intensities up to about a hundred kW/cm^2. At still higher excitation the two dimensional electron-hole plasma (EHP) related to the "Mott transition" appears. This is the level at which optically pumped quantum well lasers exhibit stimulated emission. The energy of the EHP is lower than that of the new excitonic state. These phenomena have been observed in structures of $Ga_{1-x}Al_xAs$-GaAs quantum wells. Studies also included the observation of the luminescence in high magnetic fields. The most spectacular results were those of the Zeeman spectra observed in a single quantum well. The theoretical analysis accounts

for the quantitative behavior of the complex 2D spectra of this narrow line structure.

In addition to these investigations, the study focussed on the optical properties of semiconductor devices. The first of these involved the behavior of junction diode lasers in which the active region consisted of either single or multiple quantum well lead telluride lasers. The spectral structure exhibited unexpected anomalies as a function of excitation levels. At higher intensities the spectral structure displayed transitions which have been accounted for by second order perturbation predicting the breakdown of the usual selection rules for interband transitions. The more complicated magneto-optical spectra can be similarly accounted for by the same theoretical approach. Another device which has been conceived is a periodic heterostructure laser in which the active medium in GaAs determines the frequency in conjunction with the periodicity. This has been analyzed by a transfer matrix method leading to two complex simultaneous transcendental equations. As the material technology evolves, such a periodic structure should have advantages over the passive distributed feedback laser used at present.

The third class of devices utilizing the unique properties of multiple quantum wells is the HgTe-CdTe structure. This has been postulated by McGill as an alternative to the mercury cadmium telluride alloys which are used in the long wavelength infrared at present. The important question we addressed was that of the absorption coefficient. Using $k \cdot p$ theory and actual values of masses measured in HgTe of holes

and electrons an estimate was made of the absorption coefficient in
Q-W structures and compared to those of the alloy of comparable
energy gap. The estimates indicate a qualitative agreement with McGill's
calculations but are somewhat smaller than his numbers by a factor of
three. Although luminescence spectra of the energy levels in such
structures exist, no experimental results are available of absorption
to test the validity of either theoretical estimates. Such a comparison
between experimental and theory is crucial for the justification of
fabricating this new class of infrared detectors.

II. Cumulative List of Publications

84-K-0431-1   "Exciton Transport in Optically Excited $Al_xGa_{1-x}As$-GaAs
Single Quantum Well", H.Q. Le, B. Lax, P.A. Maki,
S.C. Palmateer and L.F. Eastman, J. Appl. Phys. **55**,
4367 (1984).

84-K-0431-2   "Exciton Transport in Optically Excited $Al_xGa_{1-x}As$-GaAs
Heterostructures", H.Q. Le, B. Lax, P.A. Maki,
S.C. Palmateer and L.F. Eastman, Bull. Am. Phys. Soc.
29, 257 (1984).

84-K-0431-3   "Zeeman Structure of Excitons in GaAs-AlGaAs Single
Quantum Wells", B. Lax, H.Q. Le, P.A. Maki,
S.C. Palmateer and L.F. Eastman, Bull. Am. Phys.
Soc. **29**, 256 (1984).

84-K-0431-4   "Magneto-optical Studies of 2D Electron-Hole Plasma

in Al/GaAs Multiquantum Wells", B. Lax and H.Q. Le,

Bull. Am. Phys. Soc. 30, 382 (1985).

84-K-0431-5   "A New Quantum State in Multiple Quantum Well

Structures", H.Q. Le, B. Lax, B.A. Vojak, A.R. Calawa

and W.D. Goodhue, Proc. of the 17th Intl. Conf. on the

Physics of Semiconductors (Springer-Verlag, New York)

1985, p. 515-518.

84-K-0431-6   "High-density Excitonic State in Two-dimensional

Multiquantum Wells (accepted for publication in Physics

Review B/Rapid Communications).

III. Copies of Preprints/Reprints

Exciton transport in optically excited $Al_xGa_{1-x}As$-GaAs single quantum well

H. Q. Le and B. Lax[a]

Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
and
Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Massachusetts 02173

P. A. Maki, S. C. Palmateer, and L. F. Eastman
School of Electrical Engineering, Cornell University, Ithaca, New York, 14853

Evidence of exciton transport in optically excited $Al_{0.2}Ga_{0.8}As$-GaAs quantum well structures at low temperature and low excitation intensity is demonstrated. Photoluminescence studies of single quantum wells showed an increase in the luminescence intensity of free excitons in the GaAs quantum well with a corresponding decrease in the exciton luminescence of $Al_{0.2}Ga_{0.8}As$ barriers as temperature is varied from 1.9 to 16 K. Magnetic fields up to 100 kOe were applied in order to hinder ambipolar transport. It was found that magnetic fields caused no effect on exciton luminescences. These results indicate that excitons are formed in the barrier domains and migrate to the quantum well. Further analysis leads to the interpretation that excitons undergo relaxation without being dissociated in a quantum well. It was found that a thermal activation energy $E \sim 3.5 \pm 0.5$ meV is involved in the transport of excitons. The origin of this activation energy is considered.

PACS numbers: 78.55.Dr, 71.35. + z

I. INTRODUCTION

Much of the general interest in superlattices and quantum wells is focused on the physical phenomena that occur within the active domains, i.e., the lower-gap materials. This is particularly true for studies of such optical properties of heterostructures as photoluminescence or stimulated emission in quantum wells. There is also considerable interest in the bulk optical properties of the higher-gap materials, such as $Al_xGa_{1-x}As$, which constitutes the inactive domains of heterostructures. However, there have been fewer studies that are focused on the phenomena that occur across the boundary of the two materials. Such questions as the mechanism of carrier migration from the inactive domain to the active domain have not received much attention. This study is a contribution to the examination of this phenomenon. In particular, various aspects of exciton transport are our principal concern.

It is generally assumed that electron-hole pairs created in a semiconductor by the absorption of photons with energy high above the band gap quickly lose their initial correlation and dissociate into a two-component plasma.[1-3] The excitonic effect is known to be significant near the band gap,[4] but is reduced in strength at higher excitation energies and higher carrier densities.[5] The time scale for the dissociation is estimated to be on the order of 10^{-11}–10^{-10} s for GaAs.[2] These electrons and holes dissipate the excess energy via LO phonon emission and thermalize to states at the extrema of the bands. Surviving excitons and those reborn from the plasma serve as a major channel for electron-hole recombination; this fact is evidenced from numerous photoluminescence studies.[6] The balance between these three species,

namely, free electrons, holes, and excitons, in photoexcited bulk GaAs has been investigated to some extent,[3] but the results are at best qualitative. The difficulty in these studies does not just lie in the experimental techniques, but also in the fact that the interactive dynamics among the three species depend on such extrinsic conditions as impurity concentration, excitation intensity, and is governed by such complex intrinsic processes as electron-electron, electron-hole, and electron-phonon scattering. In addition to the problem of local equilibrium, there is also a question concerning the spatial distribution of carriers. If the carrier mean free path is small in comparison with the diffusion length and the absorption length, the spatial distribution can be understood in terms of diffusion models.[7,8] Most heterostructures have dimension comparable with a typical diffusion length, ~ 1 μm (Ref. 8) or less, and thus diffusion can still be a useful concept.

The scenario of the photoexcited semiconductor described above is even more intricate in a heterostructure. In these structures where there are two domains with different band gaps, optically generated carriers (electrons and holes) in the higher-gap domains ($Al_xGa_{1-x}As$) drift into the lower-gap domains (GaAs) which serve as sinks for the carriers. The process of relaxation and trapping of hot electrons in a quantum well has been considered in studies of quantum well lasers.[9] These electrons, which occupy states with energy at the top of the quantum well, undergo fast relaxation via LO phonon emission, 10^{-12} s, and ultimately settle at the lowest band. Evidence of these high-energy electrons is revealed in the luminescence radiation whose energy corresponds to the recombination of such electrons with holes at the top of the valence band quantum well (valence QW inverted). The relaxation of holes was found to be extremely efficient in these studies. In order to assess the efficiency of

[a] Also Physics Department.

the electron relaxation mechanism, Holonyak et al.[9] employed Fermi's "age theory" to determine the energy distribution of electrons after traveling a distance L, given a certain initial distribution. Understandably, it was found that a thicker well has a higher probability to trap electrons. These experiments were conducted at ~77 K and at high excitation intensities, ~10^4 W/cm^2.

Our present study, which was performed at lower temperature and lower excitation intensities, suggests that exciton, or correlated electron-hole pair transport, is a major participant in the process whereby carriers are collected in the active layer. Excitons have been observed to undergo relaxation without being dissociated. The evidence supporting this interpretation is derived from photoluminescence studies of single quantum well structures.

Photoluminescence spectra are studied as functions of temperature. The relative variation between the luminescence of the barrier domains (Al$_x$Ga$_{1-x}$As) and that of the quantum well (GaAs) is interpreted as the manifestation of the dynamics of carrier distribution between the two. To the extent that a diffusion model is valid, a magnetic field is applied to hinder the motions of charged particles while leaving the transport of neutral particles unaffected. The effects of a magnetic field on various luminescence lines then help to discriminate and confirm the interpretation of the origins of these lines. A model is employed to explain the quantitative aspect of the luminescence variations as functions of temperature. Detailed analysis of the spectrum of quantum well luminescence leads to the hypothesis that excitons undergo relaxation without dissociation. Qualitative theoretical consideration renders support to this proposed view.

In Sec. II, we briefly describe the experimental apparatus and procedure. Results are presented in Sec. III and the interpretation is given in Sec. IV.

II. EXPERIMENT

The heterostructures in our study consist of a single quantum well of width ~160 Å straddled by two Al$_x$Ga$_{1-x}$As barriers with thicknesses of 0.5 and 1.0 μm. The configuration of a typical sample is shown in Fig. 1. The samples are undoped and slightly p typed. We conducted experiments on two samples, 638(A) and 638(N), which have the same quantum well width, and the same mole fraction $x = 0.2$. Details of sample preparation are described by Maki et al.[10]

The experimental apparatus is standard for photoluminescence studies.[6] A 15-mW HeNe laser is used to excite the sample which is mounted strain-free in a dewar with a variable temperature control (from 1.4 to 100 K). The laser beam is focused to a spot about 80 μm in diameter, and is attenuated with neutral density filters. The luminescence is detected in the backward emission configuration, and is normal to the sample layers. The light is analyzed with a 0.75-m double grating spectrometer, and detected with a cooled GaAs cathode photomultiplier. The luminescence spectrum is digitized and integrated by numerical summation. Since the desired quantity is actually the photon flux rather than the radiation power of the luminescence, the photon count-
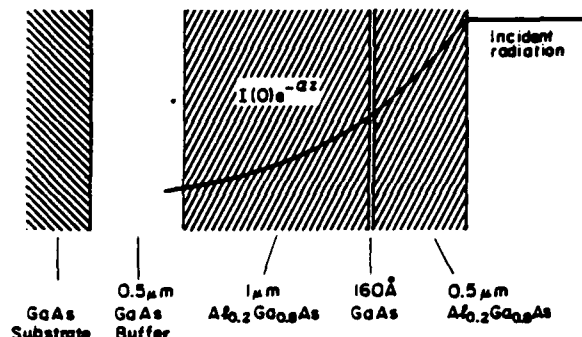

FIG. 1. Structure of a typical sample. The plotted curve is the absorption profile in the sample.

ing rates at different wavelengths are scaled with respect to the estimated quantum efficiency of the detecting system. This turned out to be only a minor adjustment, ~10%. The statistical error of the integrated intensity of a spectral line is determined by measuring the same spectral line N times (about 10), and the average value assumes an error equal to the standard of deviation divided by $\sqrt{N}$. The source of stochastic fluctuation is not in the intensity of the emission, but is due to the optical detection setup. The error in the measurement of the integrated intensity of a single line is about 1%, or better. Temperature is determined with relative uncertainty $\approx 10\%$. The sample is also placed inside a Bitter magnet which provides a field up to 100 kG.

III. RESULT

Various photoluminescence spectra were obtained as temperature T was varied from 1.9 to 16 K. The experiments were conducted at three different values of excitation intensity, 0.05, 0.5, and 1 W/cm^2; the lowest value produced the most useful results. We reproduced very similar results for both samples, 638(A) and 638(N). In the following, we first present our interpretation of the luminescence spectra, and subsequently describe their variations with respect to temperature. The dependence on magnetic field is presented along with the discussion in Sec. IV.

A. Identification of spectral lines

The evidence of a state of exciton or carrier is inferred from the luminescence corresponding to such a state. We assume that the integrated intensity of a luminescence line is proportional to the occupation number of that state, and ignore the effect of reabsorption.[6]

A typical photoluminescence spectrum of the samples consists of two sets of lines [Fig. 2(a)]. A higher-energy set, occurring at 1.78 eV [Fig. 2(c)] can be identified as the recombinative radiation from the high-gap domains, Al$_x$Ga$_{1-x}$As regions. The lower-energy set occurs around 1.53 eV and is readily identified as the luminescence from the quantum well, shifted above the bulk band gap (1.520 eV) [Fig. 2(b)].

The luminescence from Al$_{0.2}$Ga$_{0.8}$As is likely to be the recombination of impurity-bound excitons.[11-13] It is difficult to identify positively the origin of this luminescence since
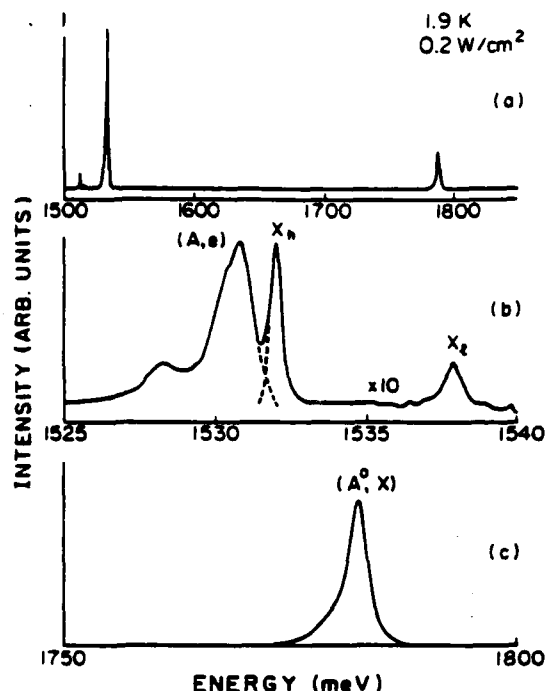
FIG. 2. (a) A broad scan of the spectrum shows the two main sets of lines. The much weaker set which occurs around 1.51 eV is the luminescence from the GaAs buffer layer. Its integrated intensity is much smaller than its height indicates. (b) A higher resolution scan of the luminescence from the quantum well. The lines marked with X_h and X_l are, respectively, the heavy-hole exciton and the light-hole exciton emissions. Other lines are impurity states. (c) The luminescence from $Al_{0.2}Ga_{0.8}As$.

there is considerable difference among published results, probably due to samples made by different growth techniques. The impurity states of $Al_{0.2}Ga_{0.8}As$ are nevertheless similar to those in bulk GaAs, which are quite well known. From a buffer layer of GaAs in the samples (Fig. 1) we obtain information about the $Al_{0.2}Ga_{0.8}As$ luminescence. The luminescence from the buffer, though quite weak, displays two main features, commonly assigned to neutral acceptor bound exciton transitions (A^0,X)(at 1.512 eV) and band-acceptor or donor-acceptor pairs (at 1.494 eV).[6,14] The former is much stronger. From this result we infer that the observed line from $Al_xGa_{1-x}As$ is most likely associated with (A^0,X). There is also a much weaker and broader line from $Al_xGa_{1-x}As$ with lower energy (1.74 eV) which is probably associated with a carbon center.[13]

The quantum well luminescence can be further separated into two subsets [Fig. 2(b)]. A higher-energy set with two peaks is intrinsic and identified as the free exciton recombination emission associated with the light hole and the heavy hole. The existence of free exciton luminescence in a quantum well is well documented,[15,16] and has been identified previously for our samples.[10] The lower-energy set is extrinsic, similar to the spectrum of p-doped quantum well structure.[16] These lines appear to be mixtures and some components are associated with electron-acceptor states. Evidence that substantiates this speculation will be considered in Sec. IV.

It can be seen from Fig. 2(b) that the heavy-hole exciton line is well resolved from the impurity line. We can extrapolate the line shape and determine the integrated intensity of

each with reasonable accuracy. The integrated intensity of $Al_xGa_{1-x}As$ luminescence is denoted by I_b, the quantum well extrinsic luminescence by I_{imp}, and the heavy-hole exciton by I_{ex}. The luminescence of the light-hole exciton is much weaker and shall be neglected for this discussion.

The absorption length for $Al_{0.2}Ga_{0.8}As$ at 2 K and 1.959 eV (6328 Å) radiation can be interpolated from the measurement by Monemar et al.,[12] and is estimated to be ~1 μm. The corresponding absorption length for GaAs is ~0.4 μm ($\alpha = 2.5 \times 10^4$ cm^{-1}).[17] The thickness of the GaAs layer is only 160 Å whereas for $Al_{0.2}Ga_{0.8}As$, it is 1.5 μm. Thus more than 96% of absorbed light generates carriers in the $Al_{0.2}Ga_{0.8}As$. Figure 2(a) shows that the luminescence from the quantum well is more intense than the luminescence from $Al_xGa_{1-x}As$. This suggests a relatively high efficiency of the quantum well as a sink for carriers.

B. Temperature dependence

As temperature is varied from 1.9 to 16 K, the total integrated intensity $I_b + I_{ex} + I_{imp}$ remains roughly constant, while there is a decrease in I_b and a corresponding increase of I_{ex}, with I_{imp} effectively unchanged. The fluctuation in the value of the total sum is much smaller than the variation of either I_b or I_{ex}. The behavior of I_b, I_{ex}, and I_{imp} as functions of T is shown in Fig. 3. This same behavior is observed for the different excitation intensities mentioned above, although the quantitative details are somewhat different.

IV. DISCUSSION

The most salient feature of the data is the difference between I_b and I_{ex} as functions of T. The constancy of the total integrated intensity $I_b + I_{ex} + I_{imp}$ is consistent with the expectation that the numbers of generated carriers are constant for a fixed excitation intensity, and quantum efficiency of the sample does not vary significantly with respect to temperature. The increase of I_{ex}, the free exciton luminescence in the quantum well, is clearly at the expense of I_b, the luminescence of excitons in $Al_{0.2}Ga_{0.8}As$. The straightforward interpretation of this effect is that more carriers or excitons are collected by the well at higher temperature. The important and interesting question that remains is whether it is free electrons and holes, which escape impurity trapping in $Al_{0.2}Ga_{0.8}As$ and migrate to the well that are responsible for the observed increase of free excitons in the well, or it is the direct exciton transfer from one domain to the other. To answer these questions we examine several aspects of the experimental results.

A. Free electrons and holes hypothesis

If we suppose that only free electrons and holes enter into the quantum well, independently or from ionized excitons, then the exciton luminescence should vary quadratically as

$$I_{ex}(T) = \gamma_x [N_e(0) + \Delta N_e(T)]^2$$

$$= I_{ex}(0) + I_{ex}(0) \frac{\Delta N_e(T)}{N_e(0)} + \gamma_x \Delta N_e(T)^2 \qquad (1)$$

and impurity luminescence, which are presumably electron acceptor recombination, should vary linearly,

$$I_{imp}(T) = \gamma_{imp}[N_e(0) + \Delta N_e(T)]$$

$$= I_{imp}(0) + I_{imp}(0)\frac{\Delta N_e(T)}{N_e(0)}, \tag{2}$$

where γ_x and γ_{imp} are exciton formation rate and impurity-carrier formation rate, respectively; $N_e(0) + \Delta N_e(T)$ is the total carrier population collected in the quantum well, and

$$\Delta N_e(0) = 0. \tag{3}$$

A simple quantitative analysis shows that the absence of an increase in I_{imp} (Fig. 3) is not consistent with Eq. (2). One might attempt to save the model by suggesting a temperature dependence of γ_{imp} in such a way that the right-hand side of Eq. (2) remains constant. In this case, it would be extremely fortuitous for the effect to occur in both samples. Although it is known that the temperature of photoexcited carriers is usually higher than that of the lattice,[1-3] this difference is not very large at the level of excitation intensity in this experiment. Furthermore, the carrier population can be changed by keeping the sample at constant temperature and varying the excitation intensity. The result unequivocally shows a corresponding linear increase of I_{imp}, though slightly slower than I_{ex}. Thus the model of independent electrons and holes is hardly tenable.

It is likely, therefore, that the increase of I_{ex} indicates an increase of excitons migrating to the quantum well. Since an exciton is a neutral particle, the presence of a magnetic field should cause no effect on its transport. For charged particles, however, the effect of a magnetic field is a well-known phenomenon in plasma studies. In the following, we discuss the relevance of a magnetic field in the study of exciton transport in our samples.

B. Magnetic field effects

The profile of carrier generation density is shown in Fig. 1. It is possible to analyze the spatial profile of carriers using a diffusion model with proper boundary conditions at the quantum well and the surface. However, the model is valid only if the electron mean free path is small compared with the diffusion length. Since many parameters are unknown for our samples, only qualitative discussion will be used to interpret the experimental results.

If a carrier is separated from the quantum well by a distance z larger than the diffusion length l_D, it has a very small probability $[\sim \text{erfc}(z/\sqrt{2}l_D)]$ of reaching the well before it is lost via recombination. In a magnetic field, the diffusion coefficient $D(H)$ is reduced[18]

$$D(H) = \frac{D(0)}{1 + \omega_c^2\tau^2} \approx \frac{D(0)}{\omega_c^2\tau^2} \tag{4}$$

for

$$\omega_c\tau \gg 1,$$

and thus, the diffusion length (provided that the recombination lifetime does not change) is
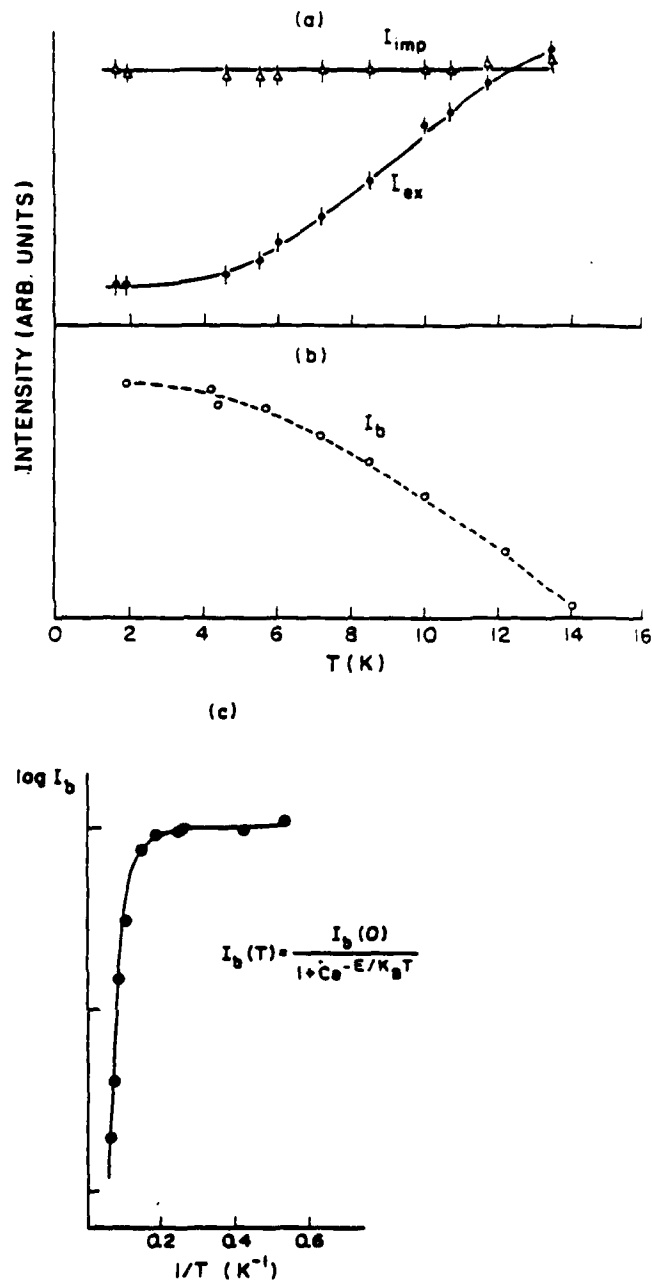
$$l_D(H) \approx \frac{l_D(0)}{\omega_c\tau}. \tag{5}$$

By applying a magnetic field, we confine carriers more tightly to their origin and thus decrease their probability of reaching the quantum well. At a field of 100 kOe, ω_c is approximately 3×10^{13} Hz. τ, which is the mean time between scattering events, is not measured for our samples, but can be estimated to be $\approx 4 \times 10^{-13}$ s from hot electron transport theory.[19] Thus $\omega_c\tau \gtrsim 10$ and l_D is reduced by at least one order of magnitude. If we use a typical figure for diffusion length in GaAs as 1 μm (Ref. 8) and assume that it is not much different for $Al_{0.2}Ga_{0.8}As$, then $l_D \sim 1000$ Å.

We conducted experiments in a magnetic field up to 100 kOe, parallel to the sample layer, i.e., normal to the diffusion path of carriers. It was found that 100 kOe produces no effect

EHP recombination close to X'. This indicates that a cold, dense system of excitons is required for the appearance of X'.

For increasing lattice temperature T_L (up to 45 K), the intensity of X' decreases while that of X_h increases or remains relatively unchanged. This relationship between X' and X_h intensities vs. T_L is also dependent on I_{ex}. This may suggest that X' and X_h represent two different phases of a dense excitonic system, in which a temperature shift causes an increase in one population at the expense of the other.

The above experimental results are applicable to a number of samples. Two samples with 145 and 63 Å well width yield the most suggestive data. Table I briefly summarizes the measured characteristics of the experimental results. There are samples which failed to exhibit this kind of below-exciton emission, instead, broad band emission extending toward higher energy, i.e. Burstein shift, was observed. We believe that in these samples, optically generated carriers fail to completely relax in energy within their radiative recombination lifetime to form a many body state.

Phenomenologically, somewhat analogous luminescence effects were observed for highly excited bulk GaAs[4,5]. A structure labeled A or P by various authors appeared to be similar to X', although the latter seems to be more pronounced. The A/P structure has been interpreted [5] as an exciton-exciton (or excitonic polariton) scattering effect. For this case, this model would involve two heavy hole excitons undergoing a collision which leaves a light hole exciton and a photon which becomes X'. However, this model does not appear to be satisfactory to account for X'. A serious objection is based on the Zeeman behavior of various states, shown in Fig. 4. The light hole exciton states split into two resolvable states with opposite polarization. Yet no such corresponding

cence in the parallel configuration. The gain factor across the excited spot was measured and found to be small for $I_{ex} \leqslant 2 \times 10^5$ w/cm^2 . The luminescence in this configuration is then essentially spontaneous. However, at higher I_{ex}, where the EHP emission appears, there is significant difference between the spectra of the two configurations obtained under identical excitation conditions, thus indicating the effect of stimulated emission on the EHP recombination. At issue in this communication is the nature of X'.

It is important to establish that X' is a different entity from the EHP. Besides the evidence from the luminescence spectra, excitation spectroscopy also contributes evidence for this distinction, as well as suggests the close conection between X' and exciton. The luminescence detected at X' displays a strong resonance with a sharp low energy edge as the excitation energy $h\nu_{ex}$ is near the heavy hole exciton (Fig. 3). This sharp edge is coincident with the low intensity CW absorption of exciton (Fig. 1 (a')). Although the bleaching of this exciton resonance eventually occurs at very high I_{ex}, it is clear that at the excitation level where X' begins to form, excitons still exist as a defined energy level of the semiconductor. This aspect is a further distinction between X' and EHP. At the electron-hole density where an EHP is formed, excitonic structure is completely bleached out[1,2]. Absorption at the intensity for EHP creation should not show strong excitonic feature. The dependence of X' on $h\nu_{ex}$ also suggests a connection between X' and exciton. As $h\nu_{ex}$ exceeds the exciton energy, X' appears to shift toward lower energy (Fig. 3, trace (b)). However, although evidences were not as obvious as in luminescence spectra, the apparent redshift of X' was probably not real, but due to the emergence of EHP in lieu of X' for $h\nu_{ex}$ larger than a certain value, at a constant I_{ex}. In summary, resonant excitation was found to be the crucial condition for the observation of a sharp and distinct X', as well as the discernment of a gradual appearance of the

solute values of I_{ex} were determined to within a factor of two, but the relative intensities were within 10%. Luminescence was analyzed with a spectrometer equipped with a GaAs cathode photomultiplier.

Luminescence spectra of emission normal to the MQW layer plane are shown in Fig. 1 for sample 1. The sample was nominally a 145 Å MQW structure as depicted in the inset. The low intensity absorption spectrum shows the two lowest exciton states, the heavy hole and the light hole exciton (Fig. 1 (a')). The large linewidths of these two lines are due to inhomogenous strains in the thin sample for absorption. Luminescence spectra at low excitation intensity (trace (a)) show the heavy hole exciton recombination (labeled X_h) as a narrower high energy peak, and other extrinsic structures at lower energy. These characteristic properties of MQW structures under low intensity excitation have been well established[3]. High intensity studies, from 10^4 to 10^6 W/cm^2 were performed using near or on resonant excitation techniques, i.e., the excitation energy $h\nu_{ex}$ was close to the ground state (heavy hole) exciton energy. Under this condition, excitons are created with very little kinetic energy. When I_{ex} exceeds 10^4 W/cm^2 (Fig. 1, traces (b)), a new feature, labeled X', emerges at about 6 meV below the heavy hole exciton. The X' linewidth is comparable to or narrower than those of the exciton states. As I_{ex} is further increased, the X' intensity grows much faster than that of X_h, and X' energy red-shifts slightly, but not very significantly. At an intensity about 30 times the threshold value at which X' emerges, a second structure appears (Fig. 2). This second structure, which rapidly overwhelms X' at still higher I_{ex}, can be presumed to be due to an electron-hole plasma (EHP), since an EHP is the ultimate limit of a high density electron-hole system. Lineshape analysis of the second structure in terms of the EHP theory yields qualitatively the expected behavior and, thus, renders support to this interpretation. The same process was observed in the lumines-

When excitons in semiconductors are present in sufficiently high density, their ultimate break-up into an electron-hole Fermi system is inevitable. However, the transitory stages between a neutral excitonic gas phase which is essentially single particle state and an electron-hole metallic phase, a many body state, is not well understood, experimentally or theoretically[1,2]. Much less understood is the problem in two dimensions. In this communication, we report the observation of luminescence spectral features from quasi-two-dimensional (Q2D) semiconductor heterostructures under high intensity optical excitation. The observed luminescence spectra bear the characteristics of many body phenomena that appear to be more complex than the two extreme phases mentioned. Using resonant excitation method to generate dense exciton systems, we studied the evolution of luminescence as function of excitation intensity. The emergence of these luminescence lines is described and their nature will be discussed.

The samples are undoped ($N_{D,A} < 10^{15} cm^{-3}$) $Al_{0.25}Ga_{0.75}As$-GaAs multi-quantum well (MQW) structures grown by molecular-beam epitaxy. Excitation was performed with an optically pumped tunable dye laser of 8 nsec pulse duration. Samples were mounted in a dewar whose temperature was varied from 1.8 to over 100 K, and in a magnetic field up to 10 tesla. The incident light was normal to the sample, and focused to a spot ≈ 100 μm. The luminescence light was detected in two configurations, either normal to or parallel with the MQW plane. In the first configuration, the luminescence is essentially spontaneous, while in the latter some stimulated emission effect may occur due to a longer active length (≈100 μm). Thus, care was taken to ensure proper experimental observation and interpretion of various luminescence spectra. Light emanating from the entire excited spot was collected, and spectra were somewhat inhomogenous. The excitation intensity I_{ex} was varied with calibrated neutral density filters. The ab-

High-density excitonic state in two-dimensional multiquantum wells*

H. Q. Le and B. Lax**

Francis Bitter National Magnet Laboratory

Massachusetts Institute of Technology, Cambridge, Ma 02139

B. A. Vojak[†] and A. R. Calawa

Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Ma 02173

ABSTRACT

A sharp photoluminescence spectral feature has been observed in AlGaAs/GaAs multi-quantum-well structures under high intensity, resonant excitation at the ground state exciton. This feature, which appears below the exciton ground state, emerges from a cold dense system of excitons, but prior to the break-up of excitons into an electron-hole plasma. A collective excitonic state is speculated.

**Also Physics Department and Lincoln Laboratory

†Present address: Amoco Research Center, P. O. Box 400, Naperville, IL 60566

IV - Acknowledgement

REFERENCES

1. C. Klingshirn and H. Haug, Phys.Rep.
70, 315 (1981).
2. J.C. Hansel, T.G. Philips and G.A.
Thomas, in Solid State Physics, ed. by
H. Ehrenreich, F. Seitz, D. Turnbull
(Academic Press, N.Y., 1977), Vol. 32,
p. 88 and T.M. Rice, ibid., p. 1.
3. For a review, see R. Dingle, in
Festkorperprobleme (Advances in Solid
State Physics), ed. by H.J. Queisser
(Pergamon-Vieweg, Braunschweig, 1975),
Vol. XV, p. 21.
4. For intrinsic luminescence properties,
see C. Weisbuch, R.C. Miller, R. Dingle,
A.C. Gossard and W. Wiegmann, Solid
State Comm. 37, 219 (1981); R.C. Miller,
D.A. Kleiman, W.A. Nordland, Jr., and
A.C. Gossard, Phys.Rev. B22, 863 (1980).
5. For extrinsic luminescence properties,
see B. Lambert, B. Deveaud, A. Regreny
and G. Talalaeff, Solid State Comm. 43,
443 (1982); R.C. Miller, A.C. Gossard,
W.T. Tsang and O. Munteanu, Phys.Rev.
B25,3871 (1982).
6. D. Olego, A. Pinczuk, A.C. Gossard
and W. Weigmann, Phys.Rev. B25, 7867
(1982).
7. A.L. Fetter, Ann.Phys. 88, 1 (1974).
8. J. Shah, Solid State Electron. 21,
43 (1978).
9. C. Benoit à la Guillaume, J.-M. Debever
and F. Salvan, Phys.Rev. 177, 567 (1969).
10. O. Hildebrand, E.O. Goebel, K.M.
Romanek, H. Weber and G. Mahler, Phys.
Rev. B17, 4775 (1978) and references
therein.
11. T. Moriya and T. Kushida, J. Phys.
Soc. Jpn. 43, 1646 (1977); ibid. 41,
849 (1976).
12. Y. Kuramoto and H. Kamimura, Jr.
Phys. Soc. Jpn. 37, 716 (1974).
13. P.K. Isihara, Y. Nakane and A. Isihara,
J. Phys. C15, 2929 (1982).
14. A. Forchel, H. Schweizer and G. Mahler,
Phys.Rev.Lett. 51, 501 (1983).
15. W.F. Brinkman and T.M. Rice, Phys.
Rev. B7, 1508 (1973); P. Vashishta,
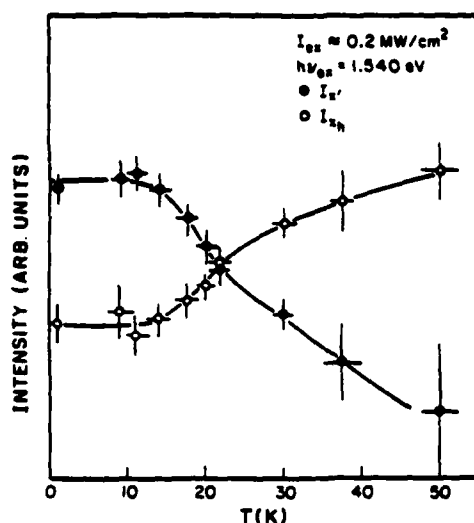S.Q. Das and K.S. Singwi, Phys.Rev. Lett.
33, 911 (1974).

Fig. 3. Temperature dependence of the integrated intensity of X' and I_{x_h}. The sample was 3-255. There was a large background at high T and X' practically vanished for T>40 K. For lower I_{ex}, X' disappeared even at lower temperature.

of the ele ron-hole pair density N_s. These parameters can be experimentally determined from the luminescence base width and the lower edge, respectively [1,2]. For a Q2D system with simple parabolic bands, c_F is linear with respect to N and E' varies as $-N^{1/2}$. Similar relations have been empirically established in bulk GaAs[10]. The observed shift and broadening of X' vs I_{ex} were too slow for these scaling relations. This argument does not necessarily preclude the existence of an EHP in MQW structures at very high I_{ex} (>0.3 MW/cm^2), but only shows that for I_{ex} from 15 to 150 kW/cm^2, X' does not seem to represent an EHP.

Further experiments are planned to investigate the possibility of a fast-diffusing nonequilibrium EHP[14], which maintains low density via diffusion.

For an EHL at a fixed temperature, c_F and E' are independent of excitation intensity since electrons and holes form droplets at an equilibrium density[2]. The somewhat stationary character of X' for a large range of I_{ex} renders the EHL interpretation a plausible candidate. If this were the case, the experimental values, shown in Table I, for various EHL parameters appear reasonable. However, our experience shows that one ought to be cautious in interpreting the linewidth of this feature since the natural linewidth may still be obscured. Furthermore, both theoretical[15] and experimental[10] investigations concluded that only EHP, not EHL is most likely to exist in bulk GaAs. Kuramoto et al.[12] and Isihara et al.[13] have treated the theoretical problem of 2-D EHL, but found no significant enhancement of favorable conditions for EHL formation with respect to the 3-D limit. For simple bands, at the limit of large hole-electron mass ratio, EHL may form in both 2D and 3D system but with a binding energy less than 10% of that of the exciton. Thus, if X' were indeed due to EHL, this would call for a careful reexamination of the theory in Q2D system.

In light of these discussions, we are led to consider the possibility of a quantum state, likely unique to Q2D system, formed by interacting excitons in the medium of an EHP. A possible boson-boson interaction, i.e., excitons exchanging acoustic plasmons is a hypothetical consideration. A system of interacting excitons appears to form a ground state prior to the formation of a degenerate electron-hole metallic phase at much higher densities. The mechanism of this interaction is under investigation.

Table I. Experimental values of various parameters if X' were due to an EHL. E' and c_F are defined in text; ϕ is the work function. N_s is given by $c_F \mu^*/\pi\hbar^2$, μ^* is the reduced mass of m_e^*=0.067, m_h^* taken to be 0.2. Also, $\pi(a_B r_s)^2 N_s$=1, a_B is the 3-D exciton Bohr radius (160 Å).

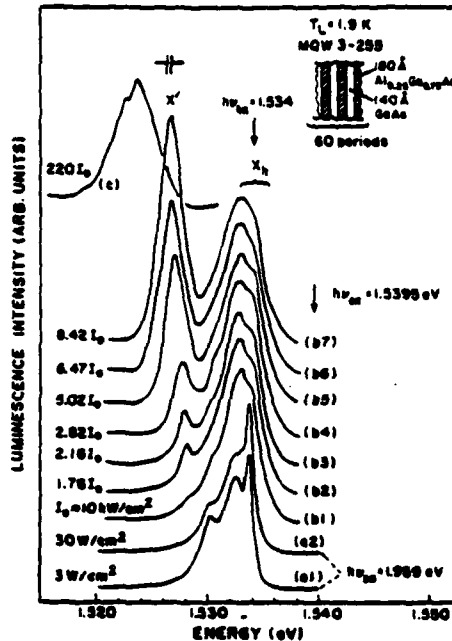| Well width	E'_g(meV)	c_F(meV)	ϕ(meV)	N_s(cm^{-2})	$r_s a_B$(Å)	r_s
140 Å	-18 ± 4	5 ± 1	-4 ± 1	1.1×10^{11}	174	1.1
68 Å	-28 ± 5	10 ± 2	-6.5±2	2.2×10^{11}	123	0.77

Fig. 1. Luminescence spectra of MQW 3-255. Traces (a) were obtained with CW low power excitation (low energy structures are impurity related which are saturated at high excitation intensity); (b) and (c) with a pulsed dye laser at 1.5395 and 1.534 eV, respectively (near or on resonance excitation). Traces (b) and (c) were smoothened; some structures are probably not real since they are well within noise level.

would be observed at lower energy if the excitation was too far off the exciton resonance. This effect can be ascribed to the high effective carrier temperature[8], which may destroy X'.

We studied the energy, lineshape, and intensity of this feature as functions of magnetic field. A magnetic field has been shown to exert significant effects on electron-hole droplets[2] when the carrier cyclotron energies are comparable to their Fermi energies. The field was applied perpendicularly to the sample layers. The quantitative results varied for different samples; nevertheless, the principal characteristics can be discerned. Generally, for low I_{ex} and near resonance excitation, the magnetic field induces a quenching effect, i.e., the intensity of the feature is reduced. This quenching effect was quite sharp in one sample (3-255). In regard to line-shape, no

significant change was observed. The Zeeman shift in energy, obtained with the lowest possible I_{ex} (near threshold), displayed exciton-like quadratic behavior (Fig. 2). This is usually interpreted as evidence of Coulomb interaction.
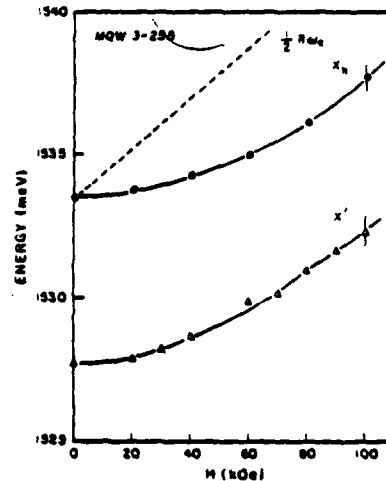

Fig. 2. Zeeman effects of X' and heavy hole exciton X_h. The dashed line represents the lowest Landau level for an electron with effective mass 0.067.

As the lattice temperature was increased, both X' and X_h red-shifted, but the change in the separation energy ΔE was smaller. At fixed I_{ex}, as the temperature was raised, I_{X_h} always increased, while $I_{X'}$ decreased. The trend is illustrated qualitatively in Fig. 3 for one value of I_{ex}.

III - Discussion

Phenomenologically, luminescence from highly excited bulk GaAs consists of a broad EHP recombination band with a shoulder labeled A or P by various authors[9-11]. This A/P structure is interpreted as an exciton-exciton (or excitonic polariton) scattering process [11]. Detailed analysis, however, shows that the feature X' observed here is at variance with this interpretation.

If X' were due to an EHP, it would be difficult to explain its sensitivity to high temperature, whereas EHP exists in bulk even for T≅77 K[10]. Furthermore, for an EHP, the Fermi energy ε_F and the renormalized band gap E'_g are rapidly varying functions

A New Quantum State in Multiple Quantum Well Structures

H.Q. Le[a], B. Lax[a,b], B.A. Vojak[b], A.R. Calawa[b], W.D. Goodhue[b]*

[a]Francis Bitter National Magnet Laboratory and [b]Lincoln Laboratory
Massachusetts Institute of Technology, Cambridge, Mass. 02139

ABSTRACT

A new luminescence spectral feature has been observed in $Al_{0.25}Ga_{0.75}As$-GaAs multiple quantum well heterostructures under selective, high intensity optical excitation. The line, which was observable only at low temperature, appears below the ground state (heavy hole) exciton line. Magnetic effects were studied. Attempts to identify this feature in terms of excitonic scattering, electron-hole plasma and electron-hole liquid suggest that the feature is a unique two-dimensional quantum state.

I - Introduction

Highly excited semiconductors have been fruitful in the research of many-body phenomena[1,2], e.g., excitonic scattering, nonequilibrium electron-hole plasma (EHP) and electron-hole liquid (EHL). In the past, both experimental and theoretical efforts have been directed primarily at the phenomena in the bulk, i.e., three-dimensional systems. In this paper, we report our observation of high density phenomena in quasi-two dimensional (Q2D) structures. A luminescence feature with energy lower than the ground state (heavy hole) exciton in selectively and highly excited $Al_xGa_{1-x}As$-GaAs multiple quantum well (MQW) heterostructures was observed. This feature, studied under different conditions of intensity, excitation energy, lattice temperature and magnetic field, displays many properties which are at variance with those models normally employed to describe high density phenomena in bulk semiconductors. In the following, we present experimental results and discuss various models. The purpose is to demonstrate some uniqueness of this feature which may represent a new quantum state.

II - Experiment and results

The sample is excited with an optically pumped tunable dye laser of 8 nsec pulse duration. The incident and luminescence radiation are normal to the sample. Temperature is variable from 1.8 to over 100 K, and magnetic fields up to 100 kOe can be applied to the sample. The samples are heterostructures grown by molecular-beam epitaxy.

Figure 1 shows a series of luminescence spectra with increasing excitation intensity of the sample depicted in the inset. The highest energy peak, labeled X_h is the ground state (heavy hole) exciton recombination; other weaker structures are impurity related. The Q2D nature of MQW structures and their luminescence properties have been well established[3-5]. As the excitation intensity (I_{ex}) exceeded 15 kW/cm^2, which corresponds to an electron-hole pair density of a closely packed exciton gas, the new feature, labeled X', emerged with relatively narrow linewidth and about 6 meV below the heavy hole exciton. At still higher I_{ex}, X' red-shifted slowly (as $\ln I_{ex}$), and a low energy tail gradually developed. The top trace of Fig. 1 corresponds to the highest excitation intensity. We designate the sharp feature observed at intermediate I_{ex} by X', and denote by ΔE the energy difference between X' and the heavy hole exciton X_h. To determine if this phenomenon is inherent to the quantum wells or contingent on the superlattice structure, we studied three other MQW samples having roughly the same quantum well width, ≈ 68 Å, but different barrier layer thicknesses. ΔE was found unambiguously the same for all these three samples, 11±2 meV. Thus, X' is not the exciton replica of superlattice plasmons[6,7] whose frequencies depend on the superlattice period. Excitation at the heavy hole exciton, detected at X' luminescence, displayed a strong resonance with a sharp leading edge, about 1 meV on the low energy side. Resonance excitation is crucial for a sharp and distinct X'. This indicates that a cold, dense gas of excitons is necessary for the formation of X'. In some samples, only a broad band

Physics and Astronomy
Classification Scheme
Number: 71.35.+z

Suggested Title of Session
in which Paper should be placed
Superlattices & Heterostructures –
Optical Properties

Magneto-optical Studies of 2D Electron-Hole Plasma
in Al/GaAs Multi-quantum Wells,* H.Q. LE and B. LAX,†
MIT Francis Bitter National Magnet Laboratory** ---
Luminescence and gain spectra of optically generated
electron-hole plasma (EHP) with densities from 10^{11} to
$10^{13}/cm^2$ per quantum well layer in AlGaAs/GaAs structures
has been studied with and without magnetic fields.
Analysis of luminescence in terms of the EHP theory[1]
showed a large band gap renormalization effect up to
$\approx$ -2 exciton rydbergs, a lineshape exhibiting the
effect of constant 2D density of states, and a
surprisingly low plasma temperature, $\leq$25K. Magnetic
fields were observed to induce a quenching effect, which
is particularly dramatic for low density EHP when the
electron cyclotron energy is comparable to the Fermi
level. Magnetic field can theoretically contain the
2D EHP expansion which has been hypothesized in 3D EHP.
Evidence for such a magnetic confinement was not
observed.

*Supported by the Office of Naval Research.
**Supported by the National Science Foundation.
†Also Physics Department, MIT.
[1]J.C. Hansel, T.G. Philips and G.A. Thomas, Solid
State Physics, ed. by H. Ehrenrich et al., (Academic
Press, NY, 1977), Vol. 32.

Prefer Standard Session

Han Le
Francis Bitter National Magnet Laborator
Massachusetts Institute of Technology
170 Albany Street, NW14-4107
Cambridge, Massachusetts 02139

Abstract Submitted

for the March Meeting of the
American Physical Society
26-30 March 1984

Suggested Title of Session
in which paper should be placed
Optical Properties of Semiconductor
Superlattices and Heterojunctions

Zeeman Structure of Excitons in GaAs-AlGaAs
Single Quantum Wells,[*] B. Lax[†] and H. Le, M.I.T.;
P.A. Maki, S.C. Palmateer and L.F. Eastman, Cornell
University --- Photoluminescence spectrum of undoped
GaAs-AlGaAs quantum well is dominated by the radiative
recombination of free excitons.[1] This salient property,
which is radically different from that of bulk GaAs,
renders photoluminescence a useful spectroscopic method
to study excitonic energy structure. Experimental photo-
luminescence studies of GaAs-AlGaAs single quantum well
in magnetic fields up to 100 kOe reveal rich spectra
containing both ground state and excited state excitons.
Quadratic Zeeman effect dominates the ground states of
light and heavy hole excitons, indicating an enhancement
of Coulomb effect due to confinement. Excited states,
observed only in high magnetic fields, can be extrapo-
lated to zero field to yield the exciton binding energy.
A theoretical model of 2-D exciton based on the
Luttinger-Koln hamiltonian will be discussed.

Prefer   Standard   Session

Benjamin Lax
M.I.T. Francis Bitter National
    Magnet Laboratory
170 Albany Street, NW14-4104
Cambridge, MA 02139

Physics and Astronomy
Classification Scheme
Number 71.35.+z

Suggested Title of Session
in which paper should be placed
Excitons and Electron-Hole Liquid

Exciton Transport in Optically Excited Al Ga$_{1-x}$ As-GaAs Heterostructures,[*] H. Le and B. Lax[†], M.I.T.; P.A. Maki, S.C. Palmateer and L.F. Eastman, Cornell University

Evidence of exciton transport
in optically excited Al$_{0.2}$Ga$_{0.8}$As-GaAs quantum well
structures at low temperature and low excitation in-
tensity will be presented. Photoluminescence studies
of single quantum wells show an increase in the
luminescence intensity of free excitons in the GaAs
quantum well with a corresponding decrease in the
exciton luminescence of Al$_{0.2}$Ga$_{0.8}$As barriers as
temperature is varied from 1.9 K to 16 K. Magnetic
fields up to 100 kOe were applied in order to hinder
ambipolar transport. It was found that magnetic fields
caused no effect on exciton luminescences. These
results indicate that excitons are formed in the
barriers domains and migrate to the quantum well where
they undergo relaxation without dissociation. A thermal
activation energy E~3.5±0.3 meV was found to be involved
in the transport. This value agrees with the estimated
exciton binding energy in Al$_{0.2}$Ga$_{0.8}$As and renders
support to the model

Prefer Standard Session

Han Le
M.I.T. Francis Bitter National
    Magnet Laboratory
170 Albany Street, NW14-4107
Cambridge, MA 02139

excitonic motion in this plane is thus not perturbed, and due to the confinement, neither is the transverse Coulomb interaction weakened.

Since the effective masses of electron and hole, and the dielectric constants are different for the two domains, their Coulomb binding energies are also different. This discrepancy in binding energy may appear in effect as an energy potential (i.e., a force field) that acts on an exciton as a whole. Excitons in $Al_x Ga_{1-x} As$ may require an activation energy to be scattered into another nonautoionizing exciton state. It is difficult, from our present experimental results, to detect and to distinguish the origin of this effect from impurity binding or the change in energy due to the traversal across the heterojunction. If the two energies are close in value, it would be virtually impossible to distinguish one from the other with this experimental approach. Other experiments are planned to investigate this effect.

E. Impurity luminescence in the quantum well

The behavior of I_{ex} and I_{imp} are not in consonnance with each other under the perturbations discussed above (temperature, magnetic field, intensity). We infer that the states corresponding to these luminescences are of different origins. Although they are separated by only 1.3 meV, which corresponds to 15 K, their thermal coupling is not clearly evidenced until higher temperature is reached (20–25 K). This means a very small value of the degeneracy of the upper state with respect to the lower state [cf. Eq. (6)] which would be implausible if the two states, a free and a bound, belong to the same quantum system. Furthermore, the quantum well impurity state appears to be an inhomogenous mixture. Theoretical calculations by Bastard[21] show that the binding energy of a hydrogenic impurity state strongly depends on the position of the impurity atom in the quantum well. Thus, the high-energy side of the impurity luminescence corresponds to impurity sites on the edge, and the low-energy tail corresponds to sites in the center of the quantum well. Since the electron-acceptor state is observed to be abundant in p-type GaAs, we belive that the observed impurity luminescence is largely an inhomogenous mixture of these states.

V. SUMMARY

In summary, this study is an investigation of transport phenomena in heterostructures. We present evidence that suggests the importance of exciton transport for electron-hole pairs in $Al_x Ga_{1-x} As$-GaAs quantum wells. The study of complementary to others which focused only on the dynamics of free carriers (electrons). Excitons are observed to dissipate energy without autoionization. This is also a peculiar property of the quantum well. Some aspects of exciton transport across a heterojunction are considered. A characteristic activation energy which affects the transport of excitons to a quantum well is found. This energy is more likely the binding energy of excitons to neutral acceptor in $Al_{0.2} Ga_{0.8} As$. However, we also have a possibility that the traversal across a heterojunction may involve an activation energy. We suggest that heterostructures can be built with

different configurations to study the statistical mechanics of excitons and carriers in the inactive domain using the active domain as a sink for sampling. In essence, the lower-gap domain can play a role similar to the alkali metal layer (Cs) coated on GaAs to study photoemission and transport in this material.[5] Since there is interest in the longitudinal transport properties of heterostructures, tunneling for example,[22] it is worthwhile to study some of these properties via optical methods.

[1]J. Shah, Solid-State Electron. 21, 43 (1978).

[2]G. R. Ulbrich, Solid-State Electron. 21, 51 (1978).

[3]C. Weisbuch, Solid-State Electron. 21, 179 (1978).

[4]See, for example, J. O. Dimmock, in *Semiconductors and Semimetals*, edited by R. K. Willardson and A. C. Beer (Academic, New York, 1967), Vol. 3, p. 259.

[5]N. F. Mott, *Metal-Insulator Transitions* (Barnes and Noble, New York, 1974), p. 149.

[6]See, for example, H. Barry Bebb and E. W. Williams, in *Semiconductors and Semimetals*, edited by R. K. Willardson and A. C. Beer (Academic, New York, 1972), Vol. 8, p. 181.

[7]See, for example, T. S. Moss, in *Semiconductors and Semimetals*, edited by R. K. Willardson and A. C. Beer (Academic, New York, 1966), Vol. 2, p. 205.

[8]L. W. James and J. L. Moll, Phys. Rev. 183, 740 (1969).

[9]N. Holonyak, Jr., R. M. Kolbas, R. D. Dupuis, and P. D. Dapkus, IEEE J. Quantum Electron. QE-16, 170 (1980).

[10]P. A. Maki, S. C. Palmateer, G. W. Wicks, L. F. Eastman, and A. R. Calawa, J. Electron. Mater. 12, 1051 (1983).

[11]J. Shah, B. I. Miller, and A. E. DiGiovanni, J. Appl. Phys. 43, 3436 (1972).

[12]B. Monemar, K. K. Shih, and G. D. Petit, J. Appl. Phys. 47, 2604 (1976).

[13]G. B. Stringfellow and R. Linnebach, J. Appl. Phys. 51, 2212 (1980).

[14]J. A. Rossi, C. M. Wolfe, and J. O. Dimmock, Phys. Rev. Lett. 25, 1614 (1970).

[15]C. Weisbuch, R. C. Miller, R. Dingle, A. C. Gossard, and W. Wiegmann, Solid State Commun. 37, 219 (1981).

[16]R. C. Miller, A. C. Gossard, W. T. Tsang, and O. Munteanu, Solid State Commun. 43, 519 (1982) and Phys. Rev. B 25, 3871 (1982).

[17]D. D. Sell and H. C. Casey, Jr., J. Appl. Phys. 46, 800 (1974).

[18]See, for example, B. Lax and J. G. Mavroides, *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1960), Vol. 11, p. 261.

[19]See, for example, E. M. Conwell, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1972), Suppl. 9, p. 105.

[20]D. Bimberg, M. Sondergeld, and E. Grobe, Phys. Rev. B 4, 3451 (1971).

[21]G. Bastard, Phys. Rev. B 24, 4714 (1981).

[22]L. Esaki and L. L. Chang, Phys. Rev. Lett. 33, 495 (1974).

on the exciton luminescence I_{ex}. There was a slight but definite decrease in I_{imp}, and the largest value of the decrease results from a low intensity experiment, about 6%. The behavior of I_b is complicated due to the Zeeman effect. It remains roughly constant. There is some noticeable increase of the carbon-center luminescence discussed above.

This experimental result is consistent with the interpretation of exciton transport. The absence of dramatic effects of the magnetic field implies that either a very large proportion of exciton transport is involved or the hindrance of the magnetic field on charged carriers is not very efficient. The truth may be a mixture of both. We note that the quantum well is located roughly at the median of the absorption profile (Fig. 1), an excellent configuration for efficient carrier collection. Thus, those electrons and holes that are generated close to the quantum well are not strongly hindered by the magnetic field. Nevertheless, the slight decrease in I_{imp} at high magnetic field suggests the presence of free electron and hole transport, and it is indeed affected by the field. The magnitude of this transport phenomenon is less certain. However, if all excitons in the quantum well were formed from independently diffusing electrons and holes, the luminescence should also demonstrate this measurable effect caused by the field. Evidently a large number of excitons in the quantum well originate from outside the well.

C. Activation model

The quantitative behavior of I_b and I_{ex} with respect to T can be explained with a simple model. If we assume that excitons in $Al_x Ga_{1-x} As$ can be in either a free state or a bound state with binding energy E, then at thermal equilibrium, Boltzmann's statistics gives

$$\frac{N_F}{N_B} = Ce^{-E/k_B T},\tag{6}$$

where N_F, N_B are, respectively, the populations of free and bound excitons. The constant C describes the relative degeneracy of the two states, which is generally large due to the higher density of free states. Let the steady decay rate of bound excitons be $\gamma_b N_B$ and that of free excitons be $\gamma_{ex} N_F$; we assume that their total creation rate is constant with respect to T,

$$\gamma_b N_B + \gamma_{ex} N_F = I_0.\tag{7}$$

It follows from Eqs. (6) and (7) that

$$I_b(T) = \gamma_b N_B = \frac{I_b(0)}{1 + C'e^{-E/k_B T}},\tag{8}$$

where

$$C' = \frac{\gamma_{ex}}{\gamma_b} C.$$

The free excitons in $Al_x Ga_{1-x} As$ evidently decay via the quantum well; thus,

$$I_{ex}(T) = I_{ex}(0) + \frac{I_0 C'e^{-E/k_B T}}{1 + C'e^{-E/k_B T}}.\tag{9}$$

Equations (8) and (9) adequately explain the experimental results.

The thermal binding energy, or the activation energy of these excitons is found to be 3.5 ± 0.5 meV for $638(N)$ and 3.8 ± 1.0 meV for $638(A)$. If the identification of $Al_x Ga_{1-x} As$ luminescence as (A^0, X) recombination is correct, this energy is the binding energy of the system

$$(A^0, X) \rightarrow A^0 + X - E.\tag{10}$$

This result agrees with a very similar study of thermal dissociation of (A^0, X) complex in GaAs by Bimberg et al.,[20] where a binding energy of 3.1 meV was found.

The process of exciton transport from one domain to another consists of two steps. The exciton in the higher-gap domain is first scattered into a high-energy exciton state in the lower-gap domain. This short-lived state decays via LO phonon emission to the more stable ground state exciton. In the following, we shall discuss this aspect of the exciton transport phenomenon.

D. The relaxation of excitons

From the experimental results presented above, it appears that excitons scattered into the well are not ionized, i.e., dissociated into free electrons and holes. An exciton at the top of the quantum well has sufficient energy to be dissociated into an uncorrelated electron-hole pair with energy lower in the well. However, an energetically allowed process is not necessarily a favorable one. This study indicates that the excitonic character is quite persistent. Excitons dissipates energy from ~ 1.78 to ~ 1.53 eV largely without autoionization.

This fact is actually in accord with the theory of excitons. The Hamiltonian of an exciton in the presence of a one-dimensional quantum well potential can be written as

$$\frac{p_{e,z}^2}{2m_e} + \frac{p_{h,z}^2}{2m_h} + V_e(z_e) + V_h(z_h)$$
$$+ \frac{P_\perp^2}{2M_\perp} + \frac{p_\perp^2}{2\mu_\perp} + \frac{e^2}{\kappa\sqrt{\rho^2 + (z_e - z_h)^2}},\tag{11}$$

where the subscript e and h of various operators refer to the operation on electron and hole vector spaces, respectively. $V(z)$ is the one-dimensional (z-dimension) quantum well potential. $P_\perp$ and $p_\perp$ are the center of mass momentum and the relative coordinate momentum in the transverse space. Since the longitudinal motions of electron and hole occur with much higher frequencies than their transverse relative motion, $|z_e - z_h|$ can be replaced by an average value in the Coulomb term. Then, the transverse motion can be decoupled from the longitudinal, and the exciton character is retained and enhanced in the transverse space. At the top of the quantum well, where the wave functions of electron and hole are spread over both domains, the effect of the quantum well is weak and the exciton in this high-energy state is intermediate for the subsequent LO phonon emission decay.

The exact detailed mechanism of exciton relaxation is certainly complicated. We note that the wave functions of an electron and a hole in the quantum well consist of large components of k_z, i.e., longitudinal momentum. Energy can be imparted to the creation of an LO phonon only if k_z changes. Thus, the emission of an LO phonon with momentum q in the z dimension is more favorable than in the x-y plane. The

splitting of X', as stipulated by the conservation of energy, was observed. Even if X' energy could be arbitrarily correlated within a linewidth to either branch of X_ℓ, the deviation (Fig. 4) is too systematic to accept the hypothesis.

In conclusion, high intensity resonant excitation on MQW structures produces a luminescence feature arising from a cold dense exciton gas, prior to the formation of degenerate electron-hole metallic phases. There have been theoretical studies[6] which predict a first order Mott transition for a 2D Coulomb gas, directly from a neutral phase to a plasma phase. The picture from the current experimental work appears to be more complicated than this theoretical model. There appears to be an intermediate state between an exciton gas and an EHP. X' could represent a collective state formed from a system of 2D interacting excitons. A possible boson-boson interaction, i.e., excitons exchanging real or virtual acoustic plasmons at certain exciton critical density is a hypothetical consideration. Such systems of interacting excitons appear to form a ground state prior to the formation of a degenerate electron-hole phase at much higher densities.

REFERENCES

1. J. C. Hansel, T. G. Philips and G. A. Thomas, in Solid State Physics, ed. by H. Ehrenreich, F. Seitz, D. Turnbull (Academic Press, N.Y., 1977), Vol .32, p.88 and T. M. Rice, ibid., p.1.

2. C. Klingshirn and H. Haug, Phys. Rep. 70, 315 (1981).

3. For a review, see R. Dingle, in Festkorperprobleme (Advances in Solid State Physics), ed. by H. J. Queisser (Pergamon-Vieweg, Braunschweig, 1975), Vol. XV, p.21. For intrinsic luminescence properties, see e.g. R. C. Miller, D. A. Kleiman, W. A. Nordland,Jr., and A. C. Gossard, Phys. Rev. B22, 863 (1980) and for extrinsic luminescence properties, see e. g. B. Lambert, B. Deveaud, A. Regreny and G. Talalaeff, Solid State Comm. 43, 443 (1982); R. C. Miller, A. C. Gossard, W. T. Tsang and O. Munteanu, Phys. Rev. B25, 3871 (1982).

4. E. O. Goebel, K. M. Romanek, H. Weber and G. Mahler, Phys. Rev. B17, 4775 (1978) and references therein.

5. T. Moriya and T. Kushida, J. Phys. Soc. Jpn. 43, 1646 (1977) and ibid. 41, 849 (1976).

6. J. M. Kosterlitz, J. Phys. C:Solid State Phys. 10, 3753 (1977); C. Deutsch, M. Lavaud, Phys. Rev. A9, 2598 (1974).

Table I. Energy (E) and linewidth (Γ) in unit of meV of X', the heavy hole exciton X_h and the light hole exciton X_ℓ for two samples. Measurements are by optical methods.

Well Width (Å)	X'		X_h		X_ℓ	
	E	Γ	E	Γ	E	Γ
145	1527.5±1	2	1533.5±0.5	1.8	1540.0±0.5	2.5
63	1570±2	5	1582±2	5	1597±8*	10

*Broad and complex structure was observed

FIGURE CAPTION

Fig. 1

(a'): absorption spectrum of MQW 1   at low intensity. X_h and X_ℓ are the heavy hole and the light hole exciton, respectively. (a1): and (a2): luminescence spectra of emission normal to the MQW layer plane (normal configuration) obtained with low intensity, CW 6328 Å laser excitation. The highest energy peak is the heavy hole exciton, and lower energy structures are extrinsic which saturate as I_{ex} is increased. (b): normal configuration luminescence spectra obtained with pulsed dye laser excitation at 1.5395 eV (near resonance excitation), showing X' emergence. (b) were smoothed; some structures are probably not real since they are well within the noise level.

Fig. 2

Normal configuration luminescence spectra at still higher excitation intensities where a second structure, identified as due to an EHP, appears and overwhelms X'.
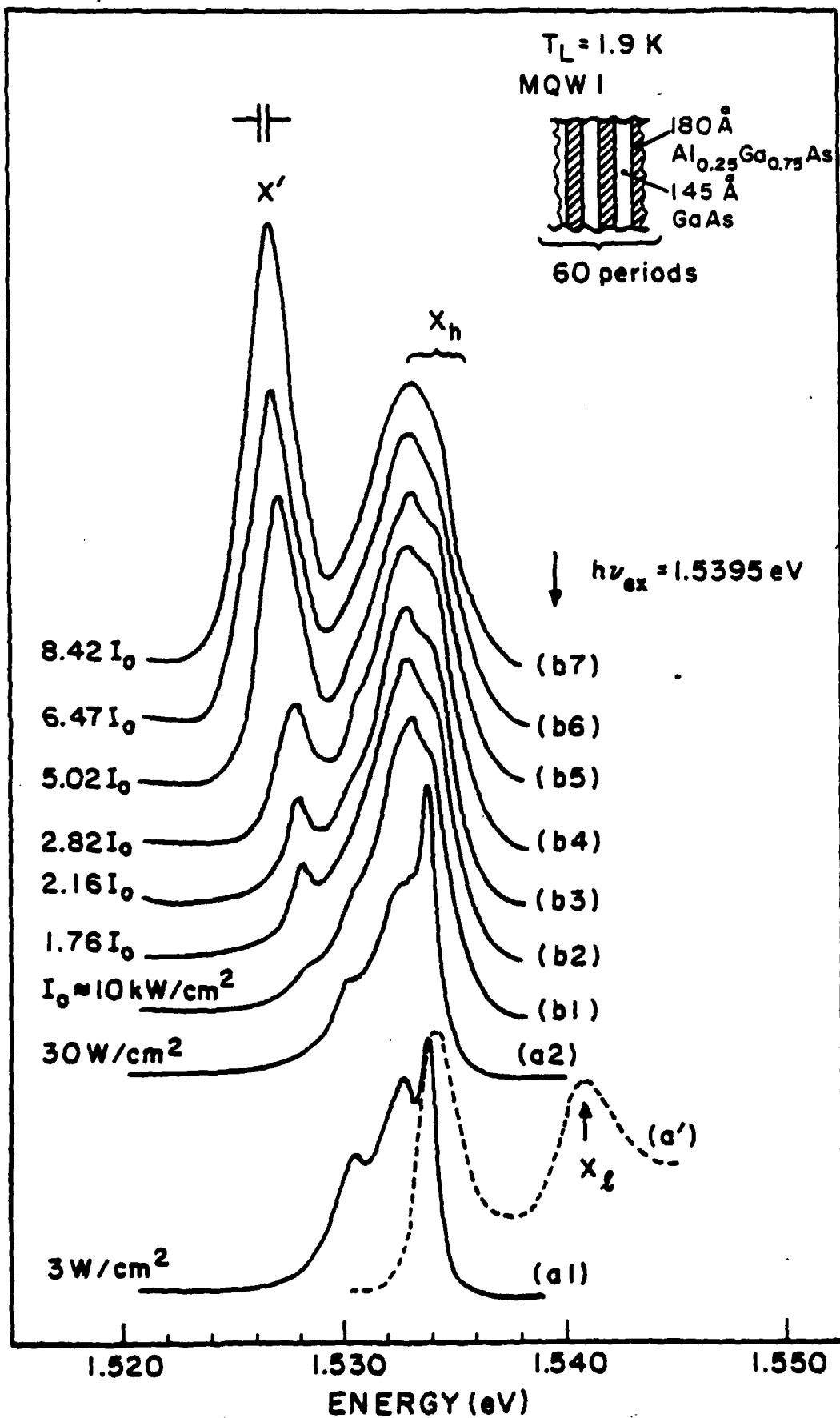
Fig. 3

(a) and (b): normal configuration luminescence spectra with $h\nu_{ex}$=1.5395 eV (near resonance) and 1.92 eV (off resonance), respectively. I_{ex} = 30±10 kW/cm^2. The structure in (b) may be due to an EHP, thus different from that in (a), which is X'. (c) and (d): excitation spectra, showing X' normal configuration luminescence intensity, collected within the indicated band (1.5 meV wide) as a function of $h\nu_{ex}$. The resonance at the heavy hole exciton X_h is also indicated by an arrow. (d) is vertically shifted for the sake of clarity.
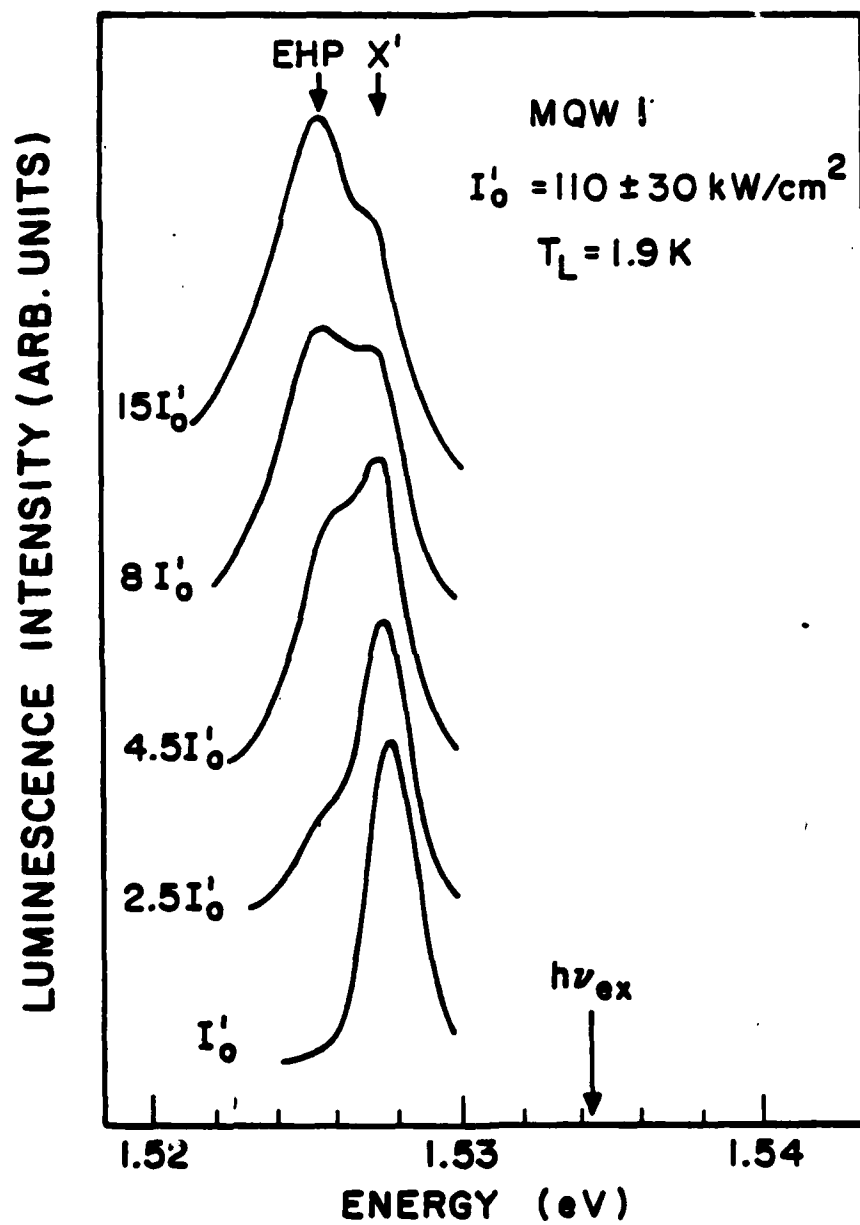
Fig. 4

Zeeman shifts of X', the heavy hole exciton X_h and light hole exciton X_ℓ in MQW 1. Data on X_ℓ were obtained via low intensity excitation spectroscopy. The labels $\pm 3/2, \pm 1/2$ represent spin quantum numbers of the hole. (a) is $2h\nu(X_h) - h\nu(X')$. According to the excitonic scattering model, conservation of energy requires (a) coincide with at least one of the two X_ℓ branches. But the deviation is clearly systematic. (b) is the theoretical calculation of free e-h pair energy.
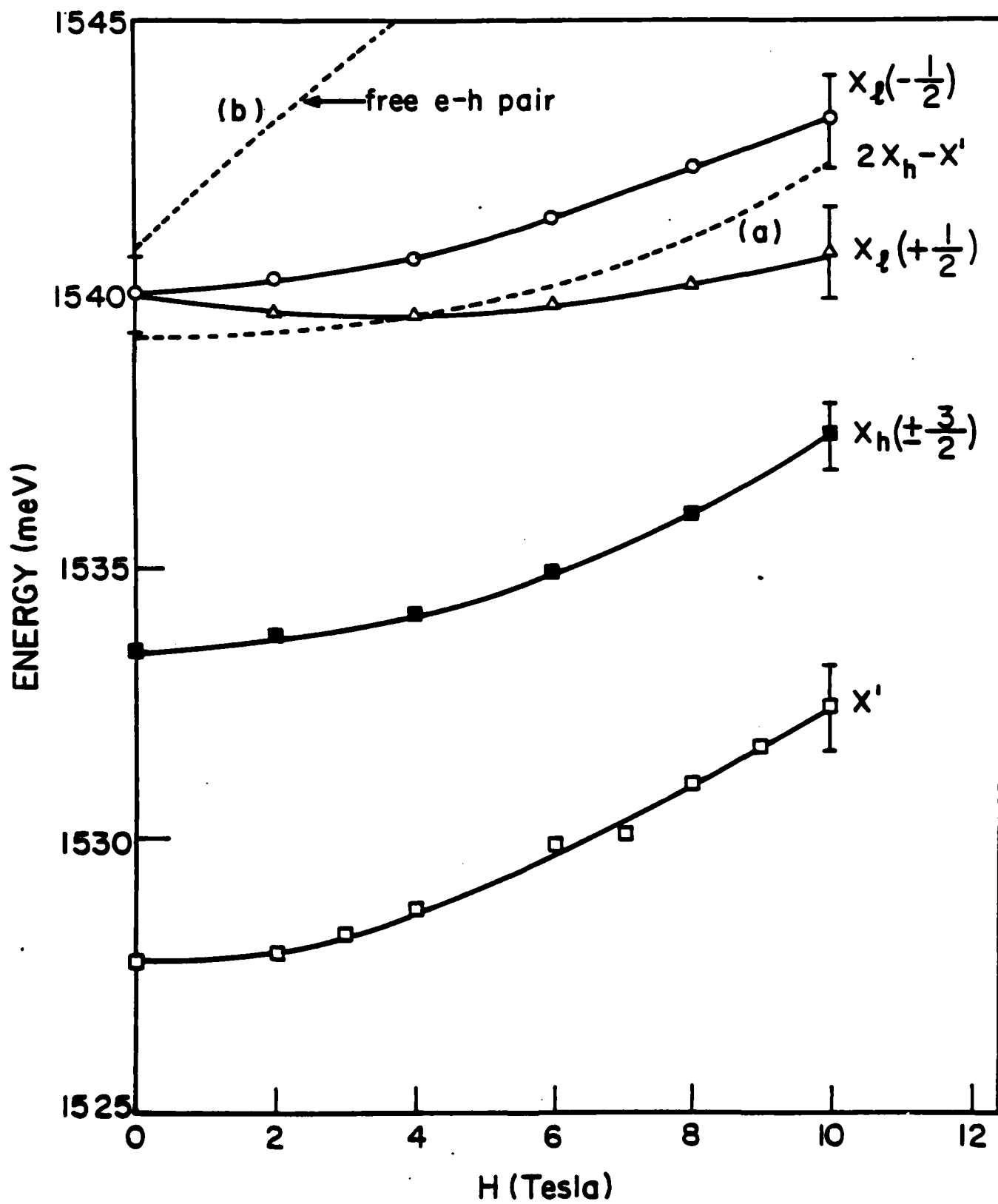
LUMINESCENCE INTENSITY (ARB. UNITS)

$T_L = 1.9K$

MQW I

ENERGY (eV)

1.520   1.530   1.540

I_0 (b)   (a)

X'

X_h

(c) I_0

(d) $5I_0$
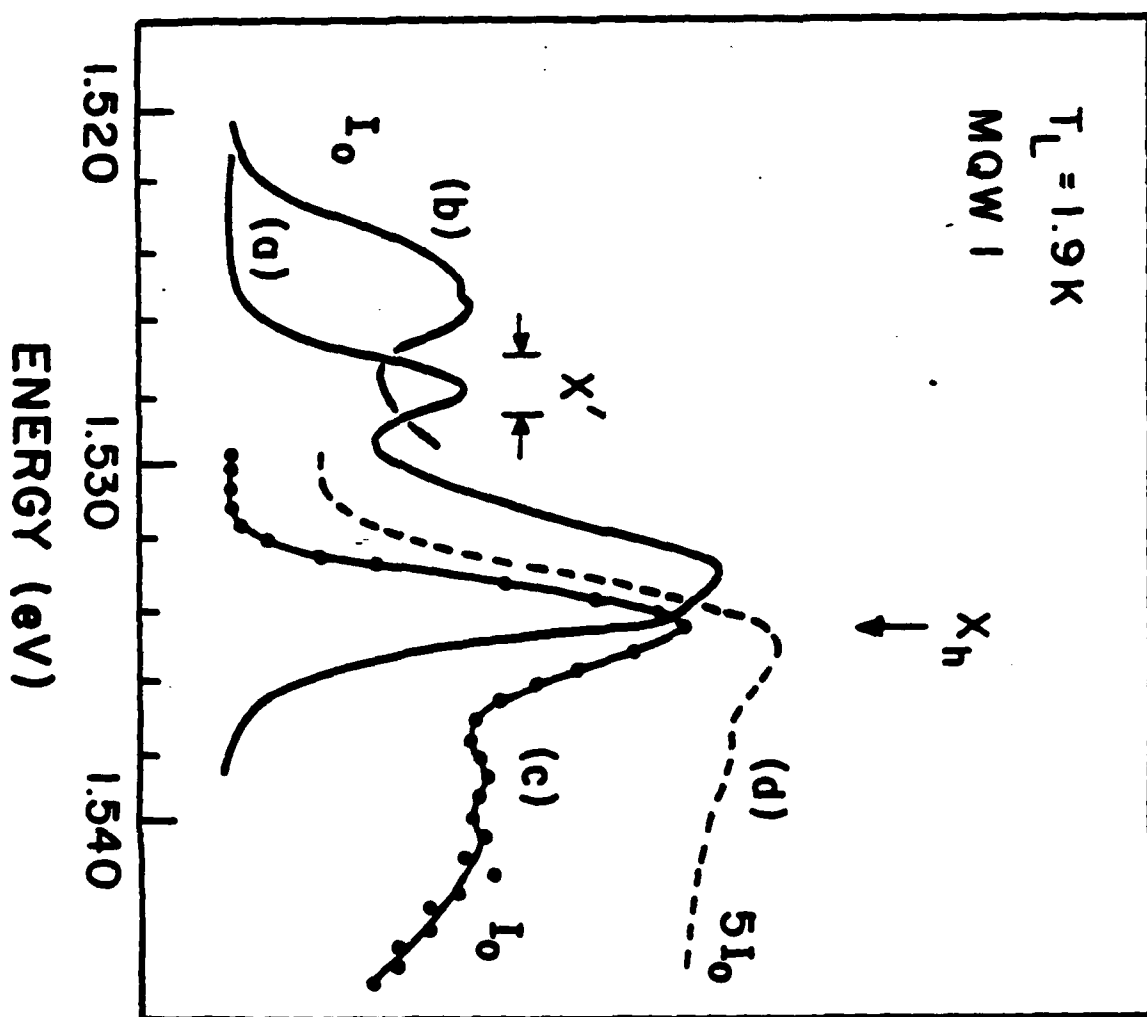
IV. List of Graduate Students/Postdocs

Graduate Students

Captain Gary Lorenzen, U.S.A.F.
          M.S., Physics, MIT, May 1984. Currently an instructor
          of Physics at U.S.A.F. Academy

Xiao-Lu Zheng

Postdoc

Dr. Han Q. Le

V. Publications/Patents/Presentations/Honors List

Papers Submitted to Refereed Journals (and not yet published)

H.Q. Le, B. Lax, B.A. Vojak, A.R. Calawa, "High-Density Excitonic States in Two-dimensional Multi-quantum Wells" (accepted for publication in Phys. Rev. B/Rapid Communications).

Papers Published in Refereed Journals

H.Q. Le, B. Lax, P.A. Maki, S.C. Palmateer, L.F. Eastman, "Excitonic Transport in Optically Excited $Al_xGa_{1-x}As$-GaAs Single Quantum Well", J. Appl. Phys. $\underline{55}$, 4367 (1984).

Contributed Presentations at Topical or Scientific/Technical Society
Conferences

H.Q. Le, B. Lax, P.A. Maki, S.C. Palmateer, L.F. Eastman, "Exciton
Transport in Optically Excited $Al_xGa_{1-x}As$-GaAs Heterostructures", Bull.
Am. Phys. Soc. 29, 257 (1984).

B. Lax, H.Q. Le, P.A. Maki, S.C. Palmateer, L.F. Eastman, "Zeeman
Structure of Excitons in GaAs-AlGaAs Single Quantum Wells", Bull. Am.
Phys. Soc. 29, 256 (1984).

H.Q. Le, B. Lax, B.A. Vojak, A.R. Calawa and W.D. Goodhue, "A New Quantum
State in Multiple Quantum Well Structures", Proc. of the 17th Intl. Conf.
on the Physics of Semiconductors, (Springer-Verlag, New York, 1985) pp. 515-518.

H.Q. Le and B. Lax, "Magneto-optical Studies of 2D Electron-Hole Plasma in
Al/GaAs Multi-quantum Wells", Bull. Am. Phys. Soc. 30, 382 (1985).

VI. Equipment

Up to this point in time, we have not spent any money on
equipment. Presently we are in the process of compiling a list of
equipment needed to carry out our research during the next year.

VII. Industrial Contacts

The principal investigator consults and collaborates with the Amoco Research Laboratory at Naperville, Illinois. This group is interested in quantum well lasers and in the evaluation of their materials grown by MBE techniques. They provide GaAs-GaAlAs samples which we use in our optical and magneto-optical studies. There is collaboration on the theoretical aspects of these investigations as well.

The principal investigator also has a similar arrangement with the General Motors Research Laboratories at Warren, Michigan. This group fabricates PbTe-PbEuTe quantum well diode lasers which we have examined at the National Magnet Laboratory in high magnetic fields. These lasers also have been provided to C. Freed at Lincoln Laboratory for heterodyne studies. The spectral results have been theoretically analyzed here at MIT and interpreted in terms of the results of this theory. This information is shared with the G.M. staff on a collaborative basis. Interest in this project has also been expressed by Laser Analytics, Division of Spectra Physics, Bedford, MA, and a collaborative program on Pb salt lasers is also contemplated with them.

Our work on HgTe-CdTe quantum well studies has attracted the attention of Honeywell Electro-optics Center, Lexington, MA. An informal exchange of ideas and information has been carried on during the past year. Hopefully Honeywell will provide samples for measurement at the Magnet Laboratory.

VIII. Collaboration With Other Academic Institutions

Our work on single quantum well structures was in collaboration with the Cornell Materials Research Center at Ithaca,NY. In particular, Professor L.F. Eastman and his students P.A. Maki and S.C. Palmateer, provided the samples for our studies.

The multiple quantum well experiments were carried out in collaboration with A.R. Calawa, W.D. Goodhue and B.A. Vojak at the MIT Lincoln Laboratory. This is a joint effort in which students from MIT will participate during the coming academic year, both at Lincoln Laboratory and at the National Magnet Laboratory under the direction of the principal investigator.

ATE
LMED
–8